Two lifetimes in the thermopower of the cuprate metals

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We study the temperature-dependence of thermoelectric transport in proton-irradiated thin films of $Tl_2Ba_2CaCu_2O_{8+\delta}$. All the anomalous transport conductivities are affected by the irradiation but maintain their non-Fermi liquid forms. We note however that the anomalous temperature dependence of the thermopower scales precisely with the temperature dependent Hall coefficient. This provides strong evidence that the two relaxation rates observed to control magnetotransport are also responsible for the anomalous temperature dependence of the thermopower.

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Understanding the normal state transport properties of the cuprate superconductors remains one of the major challenges posed by these materials. A key insight has been the notion of two distinct relaxation rates controlling transport processes, which has proved to be an important unifying concept in quantifying the anomalies in magnetotransport [1]. It addresses the temperature– and impurity–dependences of the Hall coefficient [2] as well as the violation of Kohler's rule [3]. In this Letter we present evidence that the anomalous temperature dependence of the thermopower is simply related to that of the Hall coefficient. The implication is that the same two relaxation rates which govern magnetotransport are also responsible for the unusual behavior of the thermopower.

The experimental evidence for two qualitatively different relaxation rates (inverse lifetimes) in the normal state is compelling. The resistivity, ρ_{xx}, increases linearly with the temperature at optimal doping which optical measurements [4] indicate to be due to a scattering rate, Γ_{tr}, proportional to the temperature, T,

$$\Gamma_{tr} \sim \eta T \quad (\eta \sim 2) . \tag{1}$$

Doping with scattering impurities like zinc leads to an additional residual term [5] in Γ_{tr} in keeping with Matthiessen's rule for a scattering rate. Changes in the carrier concentration away from optimal doping cause the temperature dependence of Γ_{tr} to evolve in a complex manner, as does the temperature dependence of the Hall coefficient, R_H. By contrast, their ratio (ρ_{xx}/R_H)— the inverse Hall angle—maintains a remarkably robust form [2,6,7] and also shows Matthiessen's rule behavior as the impurity concentration, n_i, is changed. Anderson interpreted this as a new scattering rate, Γ_H, which governs the decay of Hall currents [1]

$$\Gamma_H = \frac{T^2}{W} + n_i b . \tag{2}$$

These two rates must appear multiplicatively in the Hall conductivity $\sigma_{xy} \sim \omega_c/\Gamma_{tr}\Gamma_H$. Optical measurements [8] seem to confirm this form for σ_{xy}. This second rate, Γ_H, also controls the magneto-resistance [3] accounting for the failure of $\Delta\rho_{xx}(B)/\rho_{xx}(0)$ to scale with B/ρ_{xx}. To date however no one has looked for empirical evidence of these two rates outside magnetotransport. Here we study the thermopower which is the other in-plane transport property which shows systematic deviations [9–11] from the usual Drude metal form.

Electric currents can be driven by both electric fields and thermal gradients,

$$j_x^e = \sigma_{xx}E_x + \beta_{xx}\nabla_x T . \tag{3}$$

The thermopower, being the electric field per unit temperature gradient under conditions of electrical isolation, is given by the ratio of the thermoelectric and electric conductivities, $S = -\beta_{xx}/\sigma_{xx}$. For the diffusion thermopower of a Drude metal we know that

$$\sigma_{xx} = \frac{\omega_p^2}{\Gamma} , \qquad \beta_{xx} = -\frac{eT\mathcal{L}_0\omega_p^2}{\epsilon_F\Gamma} , \tag{4}$$

where $\omega_p = ne^2/m$ is the plasma frequency and $\mathcal{L}_0 = \pi^2 k_B^2/3e^2$ is the Lorentz number. Thus the scattering rates normally cancel in the diffusion thermopower and $S/T \sim e\mathcal{L}_0/\epsilon_F$ is independent of temperature. In the cuprates this relation is obeyed only in the heavily overdoped regime. Here we investigate whether the anomalous temperature dependence of the thermopower could be understood within the two relaxation rate scenario. We know that Γ_{tr} is, by definition, the scattering rate controlling σ_{xx}, but the scattering rate which should appear in β_{xx} is not, *a priori*, known. However we only require the temperature dependence of the dimensionless ratio of that scattering rate with Γ_{tr}. The only such parameter one can make out of the two rates is Γ_{tr}/Γ_H so on dimensional grounds we must have

$$S/T = f[\Gamma_{tr}/\Gamma_H] = f[\mathrm{R}_H(T)/\omega_c] \ , \qquad (5)$$

i.e. that S/T is a function of the Hall number.

Of course the above analysis makes many assumptions, in particular that the scattering rates are not strongly energy dependent and the same conduction processes enter both electrothermal and galvanomagnetic conduction. An explicit realization of this scaling relation is given by the phenomenological transport equation of Coleman, Schofield and Tsvelik [12]. From symmetry considerations they argue that to discriminate between Hall and electric currents the scattering mechanism must be sensitive to the charge conjugation symmetry of the quasiparticles. Using their expressions for the transport conductivities [13] we can obtain an explicit form for the scaling function, f,

$$\frac{S(T)}{T} = \frac{e\mathcal{L}_0}{2\epsilon_F}\left[1 + \frac{\omega_p^2}{\omega_c}\mathrm{R}_H(\mathrm{T})\right] \ . \qquad (6)$$

It is expected that the coefficients in this linear relation should depend strongly on the carrier concentration.

Experimentally testing Eqs. 5 and 6 is a delicate affair because of the extreme sensitivity of the thermopower in the cuprates to very small changes of the carrier density. Small changes in p, the number of holes/Cu at the level of $\Delta p = 0.001$ are significant enough to change the entire thermopower curve [10]. In principle, Eqs. 5 and 6 could be studied using a series of Zn– or Ni–doped samples to vary the scattering lifetimes. It would, however, be nearly impossible to ascertain the oxygen-content of the various chemically-doped samples with sufficient accuracy to keep fixed the coefficients in Eq. 6. Measurements made on a series of chemically-doped samples would also be susceptible to uncertainty from any lack of reproducibility in the geometries of the electrical and thermal contacts.

Thus we see that in order to experimentally test Eq. 6, or the more general scaling form of Eq. 5, we need to fix the carrier doping and change only the scattering rates. This can be achieved by introducing point defects using proton irradiation of thin film samples. We used a commercially-available [14] thin film of $Tl_2Ba_2CaCu_2O_{8+\delta}$. $Tl_2Ba_2CaCu_2O_{8+\delta}$ is particularly well-suited for this study because it is tetragonal, with no Cu-O chain contribution to the thermoelectric power. We measured the resistivity, thermopower, Hall effect, and Nernst effect [15] for temperatures ranging from T_c to 400 K in the as-grown sample. The sample was subsequently irradiated with 400 keV protons to a dose of 10^{16} p/cm^2, creating a uniform density of point-defects throughout the 1 μm thick sample. The sample was then re-measured with the same contacts, thermocouples and heater intact from the first measurement. It was possible using the irradiation in this way to change the scattering lifetimes without affecting either the sample stoichiometry or the contact geometries.
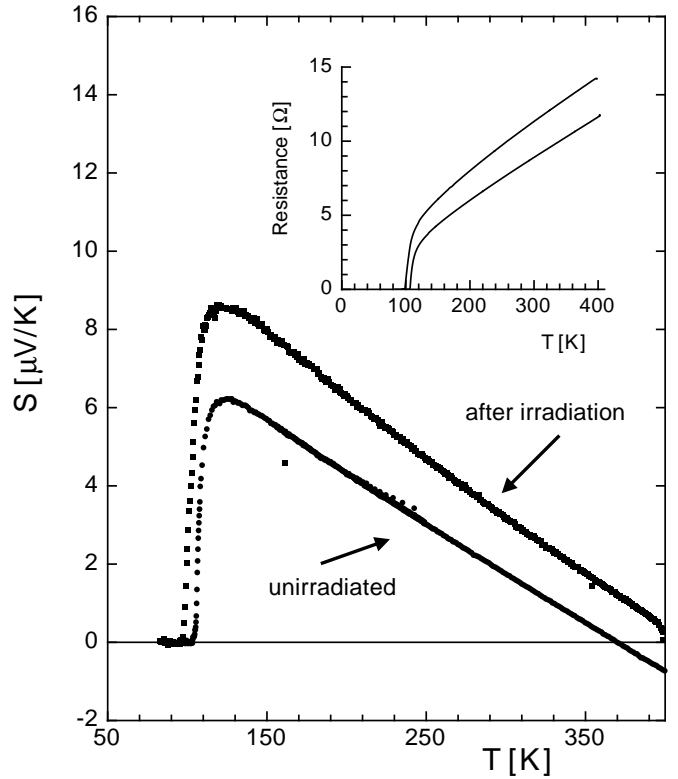

FIG. 1. The thermopower before and after proton irradiation. As discussed in the text, the proton bombardment not only increased the residual resistivity as expected, but also had a dramatic effect on the thermopower but without changing the carrier doping.

Results are shown in Fig. 1. The inset shows the resistance before and after irradiation with the upper curve showing the increased resistance after the proton bombardment. Contrary to expectation for a Fermi liquid, the irradiation had a marked effect on the thermopower, increasing it by nearly 2 μV/K. As discussed below, the upward shift of the thermopower curve appears to be an intrinsic lifetime effect, not attributable for example, to a change of carrier density from the irradiation.

Results for the Hall effect, showing the usual $1/T$ behavior, before and after irradiation are shown in Fig. 2. The Hall angle (inset) yielded a straight line when plotted against the square of the temperature both before and after irradiation. It is noteworthy that the intercept of the Hall angle curves, the residual Hall scattering, was only negligibly affected by the increased density of point defects, unlike the longitudinal residual resistance. The nature of the scatterer (point defect, Zn substitution, Ni substitution) apparently affects the two scattering channels differently.

When, according to Eqs. 5 and 6, we compare the thermopower and the Hall effect by plotting S/T as a function of R_H, the differences in the measurements before and after the proton bombardment all but vanish, as seen in Fig. 3. The two curves lie exactly on top of each other,
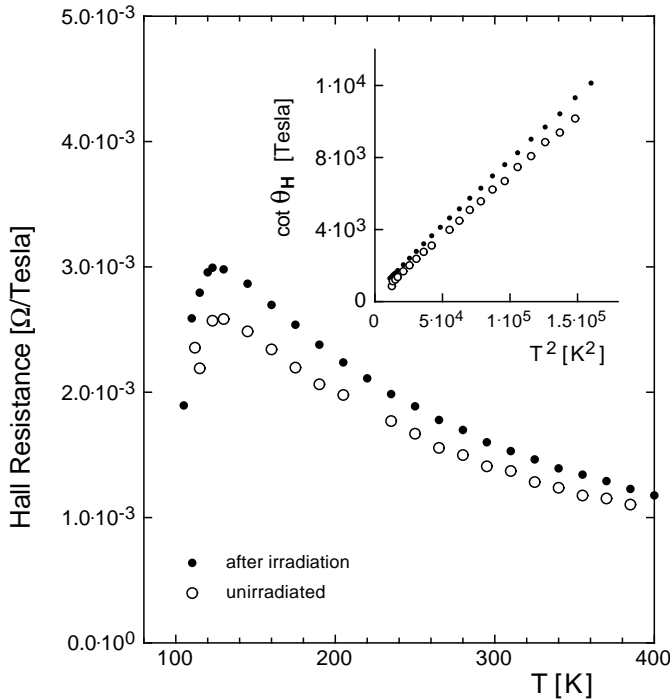
FIG. 2. The Hall resistivity and Hall angle before and after proton irradiation. The Hall angle showed the usual quadratic temperature dependence but was not very dramatically changed by proton irradiation.
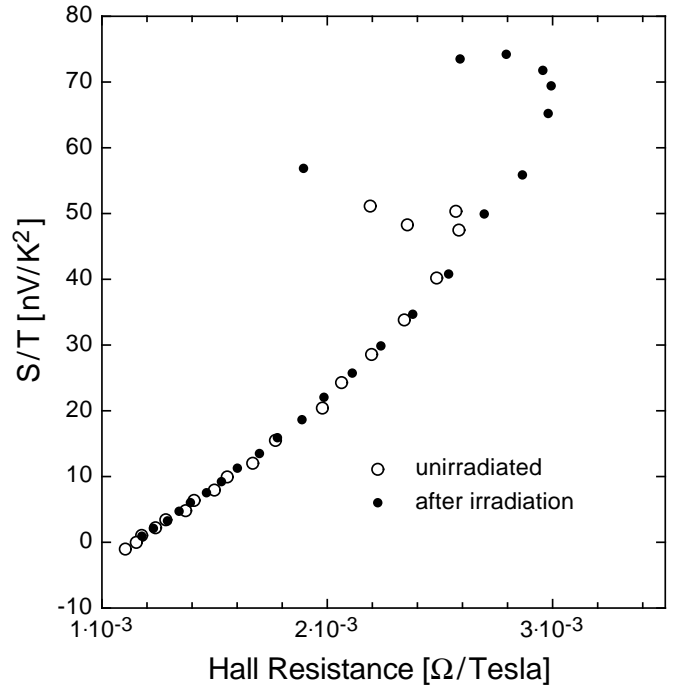

FIG. 3. Combining the results of Fig. 1 and Fig. 2 using the scaling relation of Eq. 5, we see that the curves before and after irradiation coincide. This indicates that the two relaxation rates Γ_{tr} and Γ_H determine the anomalous temperature dependence of both the Hall coefficient *and* the thermopower. Deviations occur near the superconducting fluctuation regime.

diverging only in the region of superconducting fluctuations. This is the natural consequence of Eqs. 5 and 6, in which all dependence on lifetimes is subsumed into the appearance of $R_H(T)$. The coefficients of the linear relation, Eq. 6, for example, can only depend on bandwidths, carrier concentrations, etc., but *not* the relaxation rates. Since the band structure is presumably not altered significantly by the small increase of point defects, these coefficients should be the same before and after the irradiation. This is what is observed. The slight upward curvature indicates that the general form, Eq. 5 may be more applicable at lower temperatures, nearer to T_c.

For consistency with the thermopower systematics observed by Obertelli, Cooper and Tallon [10], the slope of the curve in Fig. 3 must be extremely sensitive to changes of carrier concentration. If the proton bombardment had depleted or added carriers, the agreement of the two curves in Fig. 3 would have been highly unlikely. This result lends confidence that the experimental data were sensitive only to lifetime effects, as originally intended.

We now discuss the implications for such a scaling relation in the normal state. The obvious implication is that the two scattering rates are intrinsic to the normal state and, in particular, it is not necessary to have a magnetic field in order to observe the consequences of the Hall relaxation rate. Two microscopic mechanisms which rely on the magnetic field in this way would appear to be

disfavored by this result: the skew scattering mechanism of Kotliar *et al* [16] and the gauge model picture of Lee and Lee [17]. In the former, the presence of the magnetic field invokes a new, singular, scattering mechanism. In the latter picture the spin–fermions and charge–bosons interact strongly via a fictitious transverse gauge field which effectively screens any applied magnetic field. It is only the real electrons which can couple to the magnetic field and feel the Lorentz force so the cyclotron frequency acquires a temperature dependent factor which modifies the Hall angle.

An addition to showing that the two rates are intrinsic to the normal state in the absence of a magnetic field, this experiment has implications for the mechanism that determines which scattering rate is active in a given transport experiment. It has been customary to associate Γ_{tr} with longitudinal currents: namely those which change the energy of the quasiparticle distribution (like the current driven by an electric field). The Hall relaxation rate has been associated with transverse currents which involve a rotation of the quasiparticle distribution without a change of energy (like the Hall component of the current). An example of such a parameterization is achieved by replacing $\omega_c \tau$ by $\omega_c \tau_H$ in the usual Drude conductivities [3]. In this experiment we are seeing evidence of the Hall relaxation rate in the thermopower which is a longi-

tudinal current. Thus the distinction between longitudinal and transverse currents is not sufficient to determine which relaxation rate should be seen experimentally.

One common feature shared by both the the Hall current and the diffusion thermo-electric current is their dependence on the inverse effective mass tensor. The Hall current is determined by the curvature of the Fermi surface—the transverse effective mass. The diffusion thermopower requires hot electrons to move faster than cold holes and so is sensitive to the longitudinal effective mass. This suggests that τ_H might well be associated with any term involving the inverse effective mass, not just ω_c.

Finally we comment on theoretical pictures which predict such a relation. Anderson originally introduced the two relaxation times based on a picture of spin–charge separation in the normal state: the two lifetimes reflecting the time taken by an electron to decay into a spinon and holon (Γ_{tr}^{-1}) and the lifetime of the spinons (Γ_H^{-1}). He has also suggested [18] that these two lifetimes might show up in the thermopower based on studies of spin-charge separation in one dimension [19]. Cooper and Carrington have suggested a connection between the thermopower and the Hall coefficient based on a picture of anisotropic scattering [20]. Our motivation for considering a scaling relation came from the two-lifetimes phenomenology of Coleman, Schofield and Tsvelik [12]. In their picture the reason Γ_H enters both the Hall conductivity and the thermoelectric conductivity is indeed related to their dependence on the inverse effective mass tensor. (Currents proportional to the inverse effective mass transform under charge conjugation with the opposite sign to currents depending only on Fermi velocity.) The scaling form we find in Fig. 3 is very close to the linear relation that their phenomenology would predict (Eq. 6). However, there is one discrepancy, namely the relative sign between the two scattering rates. Their phenomenology predicts that $\beta_{xx} \sim \Gamma_{tr}^{-1} + \Gamma_H^{-1}$ *i.e.* like two fluids with thermoelectric conductivities adding. Experimentally we see that the two conductivities subtract (indicated by the positive gradient but negative intercept in Fig. 3). Thus it is as if we have two fluids which carry opposite signs of thermo-electric current. It is not clear that this discrepancy can be cured by the inclusion of a realistic Fermi surface in Ref. [12].

In summary, we have have studied the effect of proton irradiation on the in-plane normal state transport properties of $Tl2212$ thin films for a wide range of temperatures. We have demonstrated a link between the anomalous temperature dependence of the thermopower and the Hall coefficient. This suggests that the two relaxation rates used to quantify the anomalies in galvomagnetic transport are also responsible for those seen in thermoelectric transport. We have discussed the implications of this result for a number of microscopic pictures of the normal state.

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